

Toward a Universal Sea Spray Source Function (UNISOURCE)

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LONG TERM GOALS

The long-term goals of the research are to understand and assess the effects of the atmosphere on the detection of targets at low altitudes over sea in coastal regions using IR and radar systems. Effects considered are transmission losses due to aerosols and water vapor, effects of turbulent fluctuations of the air temperature on blurring and scintillation, and effects of vertical temperature and water vapor gradients on IR and rf refractivity.

OBJECTIVES

The objectives of the research performed in the framework of the present grant are:

1. To develop a source function that predicts the sea spray surface flux based on oceanographic and atmospheric parameters to within a factor of three or better independent of the geographical location, for use in chemical transport and global circulation models. Specific objectives are:
 - 1.1. Assembling an instrument package for simultaneous measurements of total and size-segregated aerosol fluxes, in conjunction with bubble spectra and relevant environmental parameters.
 - 1.2. Parameterization of sea spray fluxes as function of oceanographic and meteorological parameters.
 - 1.3. Parameterization of bubble spectra as function of oceanographic and meteorological parameters.
 - 1.4. Determination of the bubble-mediated source function at intermediate wind speeds.
 - 1.5. Determination of the relation between the bubble-mediated sea spray production and atmospheric concentrations.
 - 1.6. Determination of spume droplet contributions to the source function at elevated wind speeds and the parameterization of the spume droplet source function.
- Data from previous projects supported by ONR are analyzed and results are published when appropriate.

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APPROACH

As part of UNISOURCE, field campaigns and laboratory experiments are conducted to determine a formulation for the production of primary marine aerosol that applies in a wide variety of conditions.

WORK COMPLETED

Work completed in years prior to FY05 was summarized in earlier reports. In FY05 the following work was completed:

- Two publications using data from the ONR supported EOPACE and RED projects have been published in 2005 [De Jong et al., 2005; Reid et al., 2005]. Work on bubble and bubble plume characterization [Leifer et al.], the effects of bubbles on air-sea exchange [Woolf et al.] and work on aerosol near the surface affecting deposition of nitrogen compounds [Sørensen et al.], undertaken as part of EU funded projects and strongly related to UNISOURCE objectives, have been published FY05. Other publications listed in the publications section at the end directly result from TNO involvement in several projects through UNISOURCE: aerosol flux measurements at Mace Head [Geever et al., Heard et al.], laboratory experiments in Galway [Selegri et al.] and the UNISOURCE experiments from the FRF pier in Duck, NC in November 2004. The Mace Head experiments were a preparation for the main goals of UNISOURCE, i.e. focused measurements on sea-air aerosol fluxes at Duck, NC. See below for more detail.
- The analysis of the *laboratory experiments* at the University of Stockholm, Sweden (MISU) on bubble-mediated sea spray aerosol production under controlled conditions, using both artificial and real sea water at temperatures between 2 and 22°C, has been delayed due to sickness of the Stockholm PI. However this work has recently been continued. A new time schedule will be prepared during a meeting foreseen in the fall of 2005. The TNO bubble measurements have been analyzed and need to be interpreted in terms of water temperature and salinity, data which were recently received from Stockholm. Next the results will be analyzed in conjunction with the aerosol measurements.
- The *laboratory experiments* at the National University of Ireland in Galway (NUIG) in 2004, with the aim to study the role of organics in bubble-mediated aerosol production, have been analyzed and interpreted through comparison with field measurements at Mace Head. Results have been submitted for publication [Selegri et al.].
- Air-sea transfer studies were conducted during the *Duck2004 field campaign* from the pier of the Field Research Facility in Duck (NC, USA, see <http://www.frf.usace.army.mil/>). The aim of the campaign was to constrain the sea spray source function through a suite of complementary studies, including direct eddy covariance techniques and measurements of aerosol profiles, bubble spectra and white cap cover. Additional measurements included the air-sea transfer of momentum, heat, water vapour and CO₂. The results are analyzed in terms of ambient meteorological and oceanographic conditions. The FRF pier extends 560 m into the North Atlantic Ocean. The sea bottom gently slopes to a depth of about 10 m at the end of the pier. Supporting meteorological and oceanographic data are routinely available from the FRF.

The campaign started on November 4 and ended on November 22, 2004.

Eddy correlation measurements were conducted at the end of the pier, using methods initially developed by Nilsson et al. [1] [2]. A suite of sensors was deployed on a meteorological mast at a height of 9.30 m above the pier deck, i.e. 16.20 m above mean sea level:

- Two Solent sonic anemometers to measure the 3-D flow;
- TSI CPC 3762 to measure concentrations of particles larger than 10 nm;
- PMS optical particle counter with inlet heated at 300°C to measure the non-volatile fraction of the particle size distribution ($>0.12\ \mu\text{m}$ diam), mainly sea salt;
- CLASP (Compact Lightweight Aerosol Spectrometer Probe) based on fast response MetOne particle counters (0.2- 20 μm diameter) (from UoLeeds, Prof. Smith);
- Licor open path sampler for water vapor and CO₂ fluxes (from Colombia Univ., Dr. Zappa).

Supporting instruments included aerosol optical particle counters (PMS OAP and FSSP), an aerodynamic particle counter (TSI APS), a PMS OPC based volatility system that continuously cycled between 50°C and 700°C to determine the contributions of volatile components and sea spray to the total particle size distribution, an aethalometer to determine the absorbing aerosol fraction and a met station. Aerosol profiles were measured on a tower (SIS) with PMS optical particle counters at heights of 2, 6 and 10 m above the pier deck, which in turn was 7 m above mean sea level. The instruments were intercalibrated by operating them side by side in the tower during two periods of one full day each, at the beginning and at the end of the campaign, to ensure that they would measure the same particle size distribution. This experiment was designed for profile measurements in offshore wind, i.e. when surf-zone produced aerosol would be advected across or along the pier in a developing plume. The gradients in this plume will be used to quantitatively determine the surf zone source function [3]. A third experiment was designed to study the production of sea spray from individual breaking waves. To this end, a small float was deployed from SIS at about 20 m upwind from the pier deck with a system to measure the bubble spectra in the water, and two CLASPs at 0.5 and 1 m above the water surface. Also wind speed, temperature and relative humidity were measured on the float.

The preliminary results indicate that the conditions were representative for open water and the turbulent spectra of micrometeorological parameters and aerosol concentrations show the expected behaviour. Two periods were experienced with on-shore wind speed up to about 16 ms⁻¹, and gradually subsiding during a few days. Thus two full cycles of wind speed conditions can be analysed, with accompanying wind waves and swell. Water temperatures decreased during the campaign from about 18°C to 13°C. The profile measurements show the occurrence of gradients in offshore winds. These gradients will be used to determine the surf source strength.

Direct covariances for momentum, heat, water vapour and CO₂ are shown as function of wind speed in Figure 1, for the tow periods of high wind speed experienced in Duck2004.. These data are very preliminary and need to be corrected for various effects and further analysis and evaluation. Nevertheless, the results show the expected trends. For the aerosol fluxes the results are much less easy to interpret. Even in elevated wind speeds which favour whitecapping and thus sea spray production,

there is no clear correlation with wind speed (cf. Figure 2). This may be due to a number of reasons, e.g., correction for deposition, along shore winds producing surf produced sea spray aerosol, sea spray produced by breaking wind waves in on-shore wind. In Figure 2 no attempt has been made to separate these effects and correct for them, this is a tedious job which will be undertaken in the coming months.

The experience from Duck2004 has led to improvement of the experimental system and methodology, and the development of analysis software. This will be used in a follow-up experiment in October 2005 (Duck2005), organized by TNO (Prof. G. de Leeuw) with participation of Univ. of Leeds (Prof. Mike Smith), Colombia Univ. (Dr. Zappa) and Univ. New South Wales (Prof. M. Banner).

Preliminary results were presented at the European Aerosol Conference (EAC2005) in Ghent (B) and at the ACCENT Science Conference in Urbino (I) [De Leeuw et al., 2005].

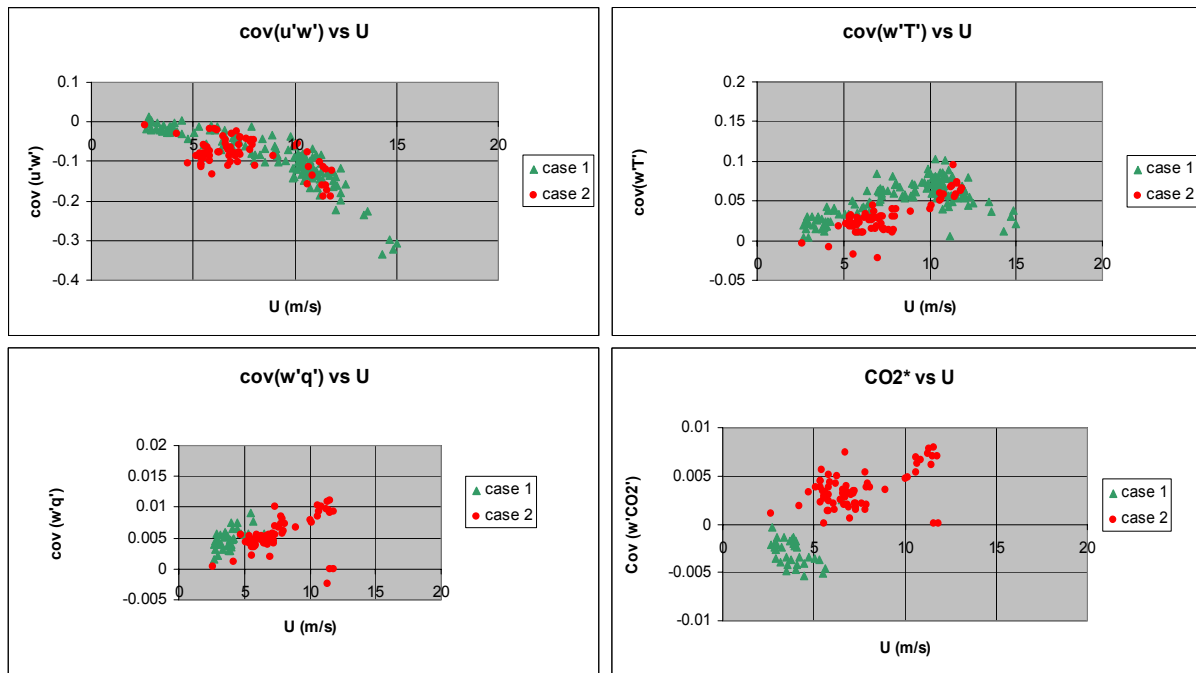


Figure 1. Direct covariances for momentum, heat, water vapor and CO_2 , respectively, vs wind speed for the two periods with high winds experienced during Duck 2005. Note that these data are very preliminary and need further analysis and corrections.

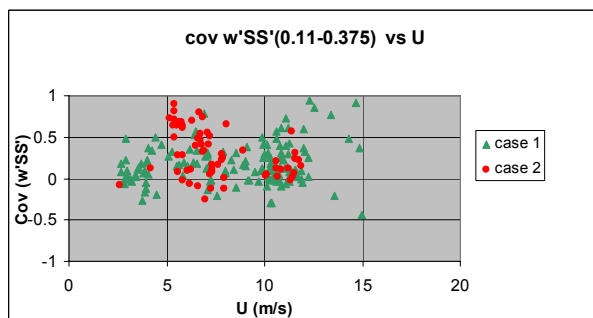


Figure 2. Direct covariances of aerosol and vertical flow velocity, plotted as function of wind speed for the two periods with high wind speeds experienced during Duck2005.

- Results from field experiments in Mace Head in 2003 are still being analyzed. A first paper has been published [Geever et al.], others are in preparation.
- Prof. De Leeuw is a member of the SOLAS (sponsored by IGBP, WCRP, SCOR and CACGP) International Steering Committee and convened a discussion session on sea spray aerosol during the SOLAS Science Conference in Halifax (Canada), 13-16 October 2004. He also convened a marine aerosol session during the European Aerosol Conference in Ghent (Belgium), 28 August – 2 September, 2005.
- During the WMO WCRP Working Group on Surface Fluxes (WGSF) meeting in Halifax, 12 October 2005, was decided that Prof. de Leeuw will lead a review of sea spray fluxes. The next SOLAS SSC will be in Amsterdam, in May 2006.

RESULTS

Results from EOPACE and RED were described in previous annual reports. The laboratory experiments indicated above have either been published or are being analyzed and results are expected early in CY06. Instrumentation including aerosol characterization equipment and a flux package is ready for use in the UNISOURCE experiments from the FRF pier in Duck, NC, in October 2005. Preliminary results from Duck2004 were reported above and presented during several scientific conferences.

IMPACT/APPLICATIONS

The results can be used to assess the effects of the atmosphere on the performance of thermal imagers over sea, and in particular the performance of LR-IRST systems. Another important application is in the field of numerical weather forecasting because of the influence on the solar irradiation at the Earth surface, as well as the assessment of the impact of aerosols on climate. Sea spray aerosol has been estimated to contribute 44% to the total aerosol optical depth, but with an uncertainty of a factor of three depending on the source function estimate used (IPCC, 2001). The surf-produced aerosol affects atmospheric processes involving sea spray particles, such as heterogeneous reactions, at fetches of at least 10 km in off-shore winds. Reaction between sea spray and HNO_3 has consequences for atmospheric inputs of nitrogen compounds in coastal waters, and thus eutrophication [De Leeuw et al., 2001a;b, 2003a;b]. Over land, sea spray influences fragile coastal eco-systems, and the corrosive properties cause damage to buildings, structures and cultural heritage. An important issue is the effect of sea spray on measurements of PM_{10} and $\text{PM}_{2.5}$. International regulation and directives on PM, e.g. by the European Commission, with frequent exceedences of the threshold values in many countries, which has severe consequences for health of the citizens and even leads to significant loss of life expectancy, has led to serious consideration of the measurements and source apportionment studies. It appears that sea salt may strongly influence the measurements of particulate mass, especially in areas close to the coast, but also further inland.

TRANSITIONS

The EOPACE and RED results of TNO-FEL are exchanged with other EOPACE and RED participants, for common analysis combining all required expertise to achieve the EOPACE and RED

goals. Common EOPACE publications have been published, others are submitted or in preparation. Accurate sea spray source functions are important in regional and global scale transport models and work is in progress with regional and global scale modelers to promote the use of our results.

RELATED PROJECTS

The efforts described above are in conjunction with other projects addressing electro-optical propagation over sea, in part basic research, in part applied research. They take place in conjunction with studies funded by the Netherlands Ministry of Defense, including work on long-range transmission, IRST and backgrounds. Data from other areas, e.g. the North Sea, the North Atlantic, the Mediterranean and the Baltic, are from other projects supported by the Netherlands Ministry of Defense, the EU or other funding agencies. Recently, EU projects have been approved to study marine aerosol production; other projects will start soon as part of the UK SOLAS programme, and proposals to the EU, in which results from, e.g., UNISOURCE will be used, are in preparation. Also in the USA there is a strong interest in marine aerosol studies, in collaboration with, e.g. TNO and UNISOURCE. Colombia Univ (Dr. Zappa) has made instrumentation available for Duck2004 and will fully participate in Duck2005.

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PUBLICATIONS

Refereed Journals:

- R.1 De Jong, A.N., G. de Leeuw, H. Winkel, P.J. Fritz and M.M. Moerman (2004). Atmospheric refraction effects on optical-infrared sensor performance in a littoral-maritime environment. *Applied Optics* 43, (34), 6293-6303. [published].
- R.2 Sørensen, L.L., S. Pryor, G. de Leeuw, and M. Schulz (2005). Flux divergence for nitric acid in the marine atmospheric surface layer. *J. Geophys. Res.* 110, D15306, doi:10.1029/2004JD005403, 12 pp. [published].
- R.3 Geever, M., C. O'Dowd, S. van Ekeren, R. Flanagan, D. Nilsson, G. de Leeuw and U. Rannik (2005). Sub-micron sea-spray fluxes. *Accepted for publication in GRL* (14 June, 2005) [published].
- R.4 Leifer, I. and G. de Leeuw (2005). Bubbles Generated from Wind-Steepened Breaking Waves: Part 1. Bubble from bubble plumes, *J. Geophys. Res. Oceans*, in press. [published]

R.5 Leifer, I., G. Caulliez and G. de Leeuw (2005). Bubbles Generated from Wind-Steepened Breaking Waves: Part 2. Bubble Plumes, Bubbles, and Wave Characteristics, *J. Geophys. Res. Oceans*, *in press*. [published]

R.6 Reid, J.S., B. Brooks, K.K. Crahan, D.A. Hegg, T.F. Eck, N. O'Neill, G. de Leeuw, K.A. Anderson, and E.A. Reid (2005). Reconciliation of coarse mode sea-salt aerosol particle size measurements and parameterizations at a sub-tropical ocean receptor site. MS 2005JD005818, *J. Geophys. Res.*, *in press*. [published]

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R.8 Selegri, K., C.D. O'Dowd, S.G. Jennings and G. de Leeuw (2005). Surfactants and sub-micron sea-spray generation. *Submitted for publication in J. Geophys. Res.* [Submitted]

R.9 Leifer, I., G. Caulliez and G. de Leeuw (2005). Characteristics of bubble plumes, bubble-plume bubbles and waves from wind steepened wave breaking. *Submitted for publication in J. Mar. Systems*. [Submitted]

R.10 Woolf, D.K., I.S. Leifer, P.D. Nightingale, T.S. Rhee, P. Bowyer, G. Caulliez, G. de Leeuw, S.E. Larsen, M. Liddicoat, J. Baker and M.O. Andreae (2005). Modelling of bubble-mediated gas transfer; fundamental principles and a laboratory test. *Submitted for publication in J. Mar. Systems*. [Submitted]

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P.1 De Leeuw, G., M.M. Moerman, M.H. Smith, S. Norris, J. Lingard, J. Gunby and C. Zappa (2005). Primary marine aerosol production studies from Duck (NC). Abstracts of the European Aerosol Conference 2005, ISBN 9080915939, abstract nr 247. [published].

P.2 De Leeuw, G., M. Moerman, S. Norris, M. Smith and C. Zappa (2005). Air-sea flux measurements and sea salt primary production. ACCENT Symposium, Urbino (I), September 12-16, 2005 [published].